

# Homogeneous linewidth of the ${}^4\text{I}_{11/2} - {}^4\text{I}_{15/2}$ optical transition of erbium in $\text{LiNbO}_3:\text{Er}^{3+}$

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**Abstract.** We work out a simple, pulsed pump-probe measurement scheme to measure the homogeneous linewidth of an atomic transition in an inhomogeneously broadened spectral line in a solid state environment. We apply the theory to the  ${}^4\text{I}_{11/2} - {}^4\text{I}_{15/2}$  optical transition of erbium in  $\text{LiNbO}_3:\text{Er}^{3+}$  crystal. Beside obtaining the homogeneous linewidth, we have estimated the population relaxation time as well.

## 1. Introduction

The successful realization of coherent quantum control procedures imposes serious requirements on the participating fields and atomic systems. The most important ones are the following: the light fields should be sufficiently coherent, the population-  $T_1$  and the dipole relaxation time  $T_2$  of the involved atomic quantum states should be sufficiently long. Experience shows that in a solid the electronic spins are the most robust quantum objects. Some of the recent achievements and feasible proposals in the field of coherent control in solids are electromagnetically induced transparency [1, 2] (exp); coherent Raman beats [3] (exp); coherent population trapping in ruby crystal at room temperature [4] (exp); slow down of light pulses in a solid [5, 6] (exp, theor) [7] (theor); quantum storage of light pulses [5, 9] (exp) [8] (theor). Coherent population transfer to the doping atoms has also been performed experimentally [10], which opens the way for the realization of quantum logic gates.

In a solid the  $T_2$  time is often orders of magnitude shorter than the  $T_1$  time, hence it is the main limiting factor for realizing coherent control experiments. The standard way of measuring the  $T_2$  time is the photon echo spectroscopy [11]. Photon echo measurements have been done for more than forty years in solids [12, 13, 14, 15, 16].

In this paper we propose an alternative technique to measure the  $T_2$  dipole relaxation time based on spectral hole burning spectroscopy [11, 17]. Our method is applicable if the atom can be treated as an effective two-level system interacting with a narrow linewidth coherent laser field. It is shown that for a sufficiently low intensity pump and probe fields, the width of the recorded absorption line-profile is proportional to the homogeneous linewidth of the spectral line. The method is applied to the transition  ${}^4\text{I}_{11/2} - {}^4\text{I}_{15/2}$  of erbium in  $\text{LiNbO}_3:\text{Er}^{3+}$ . A very detailed fluorescence spectroscopic analysis of the transition lines of  $\text{LiNbO}_3:\text{Er}^{3+}$  has been reported in [18]. The photon echo measurement of the  $T_2$  dipole relaxation time of the  ${}^4\text{I}_{13/2} - {}^4\text{I}_{15/2}$  transition has been published in [19].

The organization of the paper is as follows: in section 2 the model system is presented. The pulsed pump-probe scheme is described and the measurable absorption-line profile is derived. In section 3 the details of the experiment and the results of the measurement are presented. The results are summarized in section 4.

## 2. Model calculation

We consider a set of effective two-state atoms interacting with a long pump- and a short, read-out probe pulse. The atomic transition frequencies  $\omega_{eg}$  are spread in a range  $\mathcal{D}$ , the distribution of the atoms over this range is characterized by the function  $g(\omega_{eg})$ , where the indices  $g$  and  $e$  refer to the atomic ground and excited states, respectively. The distribution  $g(\omega_{eg})$  describes an inhomogeneous line broadening in spectroscopic language. We use the Bloch vector notation to describe the quantum state of the two-state atoms. The components of the Bloch vector  $[U_\delta(t), V_\delta(t), W_\delta(t)]_{\delta \in \mathcal{D}}^T$  can be expressed in terms of the atomic density matrix elements. They are defined as

$$U_\delta(t) = \varrho_{eg}(\delta; t) + \varrho_{ge}(\delta; t), \quad (1a)$$

$$V_\delta(t) = i[\varrho_{eg}(\delta; t) - \varrho_{ge}(\delta; t)], \quad (1b)$$

$$W_\delta(t) = \varrho_{ee}(\delta; t) - \varrho_{gg}(\delta; t), \quad (1c)$$

where the index  $\delta$  denotes the frequency difference between the transition frequency  $\omega_{eg}$  of the particular atom and some reference frequency  $\omega$ ,  $\delta = \omega_{eg} - \omega$ .

Now we consider the interaction between the atoms and a monochromatic light wave. The dynamics is described by the Bloch equations

$$\dot{U}_\delta = -\delta V_\delta - \frac{1}{T_2} U_\delta, \quad (2a)$$

$$\dot{V}_\delta = \Omega_w W_\delta + \delta U_\delta - \frac{1}{T_2} V_\delta, \quad (2b)$$

$$\dot{W}_\delta = -\Omega_w V_\delta - \frac{1}{T_1} (W_\delta + 1), \quad (2c)$$

where  $T_1$  ( $T_2$ ) denotes the population (dipole) relaxation time. The reference frequency is equal to the angular frequency of the field,  $\omega = \omega_w$ . The Rabi frequency  $\Omega_w$  is given by  $\Omega_w = -d_{eg}E_w/\hbar$ , where  $d_{eg}$  denotes the dipole moment of the atomic transition,  $E_w$  describes the pump (write  $\equiv w$ ) field strength. The pump field acts so long on the atoms that the transitions become saturated, i.e. the atoms achieve a steady state. The steady state solution to Eqs. (2) is given by

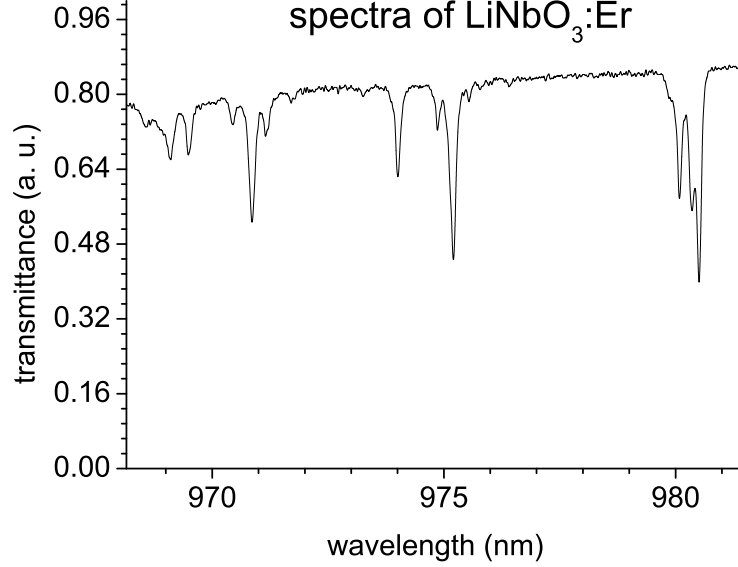
$$U_\delta(0) = \frac{\Omega_w \delta}{\delta^2 + \frac{1}{T_2^2} + \frac{T_1}{T_2} \Omega_w^2}, \quad (3a)$$

$$V_\delta(0) = -\frac{\Omega_w/T_2}{\delta^2 + \frac{1}{T_2^2} + \frac{T_1}{T_2} \Omega_w^2}, \quad (3b)$$

$$W_\delta(0) = -\frac{\delta^2 + 1/T_2^2}{\delta^2 + \frac{1}{T_2^2} + \frac{T_1}{T_2} \Omega_w^2}. \quad (3c)$$

The origin of time is set to the end of the pumping process. Hence the pumping process starts at a time instant  $t \ll 0$  and ends at  $t = 0$ . The second, probe pulse arrives after a time delay  $t_d$  that the pump pulse ceases. Assuming that  $T_2 \ll t_d$  the components of the Bloch vector are  $U_\delta(t_d) = 0$ ,  $V_\delta(t_d) = 0$ , and

$$W_\delta(t_d) = (1 + W_\delta(0))e^{-t_d/T_1} - 1. \quad (4)$$



**Figure 1.** A section of the absorption spectrum of  $\text{LiNbO}_3:\text{Er}^{3+}$ .

In rare-earth doped  $\text{LiNbO}_3$  it is generally true that  $T_2 \ll T_1$  for the ionic decay times. It is assumed that the length of the probe pulse  $\tau$  satisfies the relation  $T_2 \ll \tau \ll T_1$ . The interaction between the probe pulse and the two-state atoms are described again by Eqs. (2), where the Rabi frequencies  $\Omega_w$  should be replaced by the Rabi frequency associated with the probe pulse  $\Omega_p = -d_{eg}E_p/\hbar$ . If the above conditions hold for the time scales, and  $\tau\Omega_p \ll 1$ , then  $W_\delta(t_d + \tau) \approx W_\delta(t_d)$ , i.e. the inversion is approximately constant during the action of the probe pulse. As a result, for the coherences one obtains

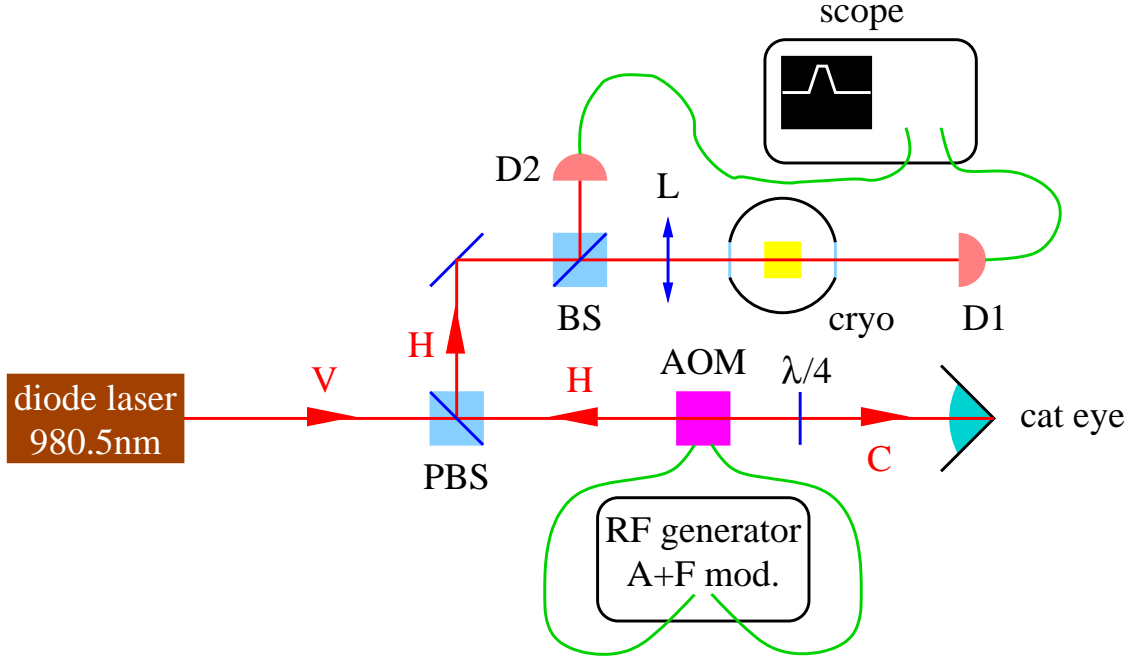
$$U_\delta(t_d + \tau) = -\frac{(\Delta - \delta)\Omega_p}{(\Delta - \delta)^2 + \frac{1}{T_2^2}}W_\delta(t_d), \quad (5a)$$

$$V_\delta(t_d + \tau) = \frac{\Omega_p/T_2}{(\Delta - \delta)^2 + \frac{1}{T_2^2}}W_\delta(t_d), \quad (5b)$$

where  $\Delta$  is the detuning of the probe pulse from the frequency of the pump (write) pulse. The susceptibility of the ensemble of the atoms is obtained from  $U_\delta(t_d + \tau)$  and  $V_\delta(t_d + \tau)$  by summing up for all subset of atoms with transition frequency difference  $\delta$ . The sum should be weighted by the distribution function  $g(\delta)$

$$\chi^+(\Delta) = -\mathcal{N}\frac{|d_{eg}|^2}{\varepsilon_0\hbar} \int g(\delta) \frac{(\Delta - \delta) + i/T_2}{(\Delta - \delta)^2 + \frac{1}{T_2^2}} W_\delta(t_d) d\delta, \quad (6)$$

where  $\mathcal{N}$  is the density of the two-state atoms participating in the process. For a wide enough distribution  $g(\delta)$ , i.e.  $\mathcal{D} \gg T_2^{-1}$ , the function  $g(\delta)$  can be brought in front of the integral. As a result, the Eq. (6) simplifies to a convolution of a Lorentzian-curve and  $W_\delta(t_d)$ . The convolution can be performed by the application of Fourier and inverse-Fourier transforms. The results is



**Figure 2.** The experimental setup for the  $T_2$  measurement. The symbols mean: BS – beam splitter; PBS – polarizing beam splitter; AOM – acousto optic modulator;  $\lambda/4$  – retarder plate; L – focusing lense; cryo – cryostat with lowest temperature of 8.2K.

given by

$$\chi^+(\Delta) = \frac{\mathcal{N}|d_{eg}|^2}{\varepsilon_0\hbar} \pi g(0) \left[ \frac{\frac{T_1}{T_2} \Omega_w^2 \Delta}{\Gamma \left( \left( \frac{1}{T_2} + \Gamma \right)^2 + \Delta^2 \right)} + i \left( 1 - \frac{\frac{T_1}{T_2} \Omega_w^2 \left( \frac{1}{T_2} + \Gamma \right)}{\Gamma \left( \left( \frac{1}{T_2} + \Gamma \right)^2 + \Delta^2 \right)} e^{-t_d/T_1} \right) \right], \quad (7)$$

where  $\Gamma^2 = \frac{1}{T_2^2} + \frac{T_1}{T_2} \Omega_w^2$ . The real part of  $\chi^+(\Delta)$  describes a phase shift for the probe pulse, whereas the imaginary part contributes to attenuation. For  $\Delta = 0$ , i.e. when the pump and probe pulses have the same frequency, the attenuation is minimal. Therefore, the imaginary part of the susceptibility in Eq. (7) describes a *spectral hole*.

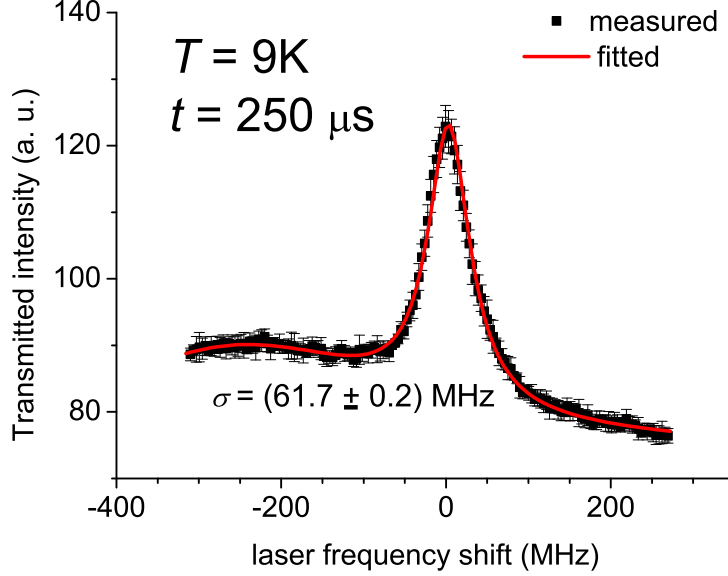
In the Eq. (7) the quantity  $(T_1/T_2) \Omega_w^2$  corresponds to power broadening. For a low intensity pump (write) pulse the result of the convolution can be expanded, in lowest order of  $\Omega_w^2$  one finds

$$\text{Im}(\chi^+(\Delta)) = \frac{\mathcal{N}|d_{eg}|^2}{\varepsilon_0\hbar} \pi g(0) \left( 1 - \frac{2 \frac{T_1}{T_2} \Omega_w^2}{\Delta^2 + \frac{4}{T_2^2}} e^{-t_d/T_1} \right). \quad (8)$$

The exponential term describes the vanishing of the spectral hole due to population relaxation. Comparing the  $\Delta$  dependence of this equation with a Lorentzian distribution  $\approx [\Delta^2 + (\sigma/2)^2]^{-1}$ , one observes that by scanning the probe field frequency through resonance with the pump field, a Lorentzian line-shape is obtained with half-width of  $\sigma = 4T_2^{-1}$ .

### 3. Experiment

We have realized the pump-probe process described in the previous section for erbium doped  $\text{LiNbO}_3$  to measure the  $T_2$  dipole relaxation time for the optical transition between the states  $^4I_{11/2} - ^4I_{15/2}$  of  $\text{Er}^{3+}$ . The absorption spectrum of a stoichiometric  $\text{LiNbO}_3:\text{Er}^{3+}$  is shown

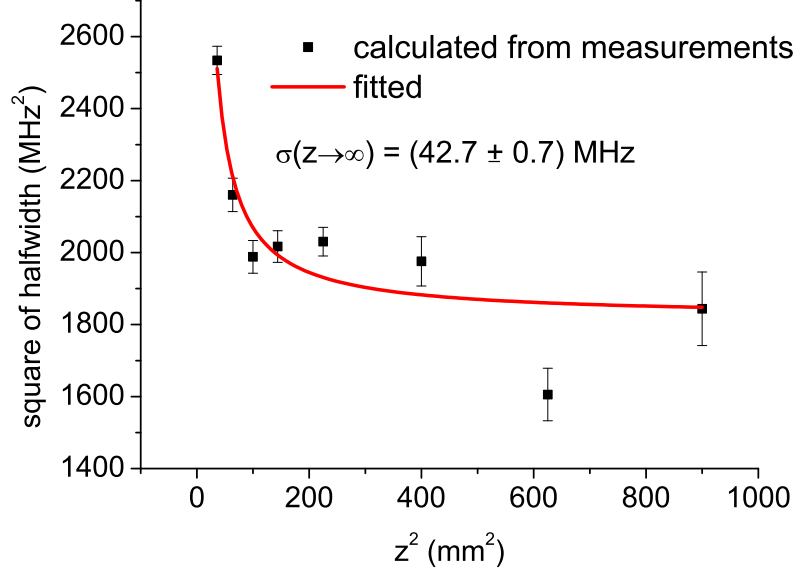


**Figure 3.** The result of several scans with the probe pulse through resonance with the pump pulse.

in figure 1. The group of lines around 980.5nm corresponds to the  ${}^4I_{11/2}-{}^4I_{15/2}$  transition. The spectral lines are incoherently broadened, the width of the deepest peak is approximately  $0.8\text{cm}^{-1} \equiv 24\text{ GHz}$ . The scheme of our experimental setup is shown in figure 2. An external cavity diode laser is tuned to the center of the peak at 980.5nm. The spectral width of the laser line is approximately 1MHz. The pump and probe pulses are derived from the same seed, the amplitude is modulated by an acousto optic modulator (AOM). In order to correct the walk-off of the diffracted beam after the AOM, a cat eye configuration is applied.

The pump pulse has a fixed frequency, whereas the frequency of the probe pulse should be scanned through the pump frequency. First we made the scanning by varying the radio frequency on the AOM. It turned out that the scanning range  $\approx 50\text{MHz}$  of the AOM is not sufficient, hence we used the built in scanning capability of the diode laser with a frequency range of several GHz. The light beam leaving the cat eye system is fed into the crystal sample, which is placed in a cryostat with lowest temperature of 8.2K. The signal and the reference beam intensity is measured by the D1 and D2 photo-diode detectors, respectively. The sample stoichiometric  $\text{LiNbO}_3:\text{Er}^{3+}$  crystal has been grown in-house by top-seeded solution growth method. The concentration of Er in the solution from which the crystal has been grown, was 0.1 mol%.

The total measurement cycle consists of three stages: a long pumping period with duration more than  $4500\mu\text{s}$ , a delay  $t_d$  of more than  $100\mu\text{s}$  when the light field is switched off, and a scanning with a probe field which takes  $200\mu\text{s}$ . The delay time is measured from the switch off of the pump field until the middle of the scan period. The time delay and the scan times are limited by the piezo-scanning rate of the diode laser head. On the other hand, the susceptibility in Eq. (7) has been derived for a fixed frequency probe pulse. Hence the rate of change of the probe field frequency should be small enough so that the system can relax with the rate  $T_2^{-1}$  to the quasi steady state determined by the probe pulse. We have verified by numerically solving the dynamical Eqs. (2) that using the above parameters the system relaxes to the quasi steady

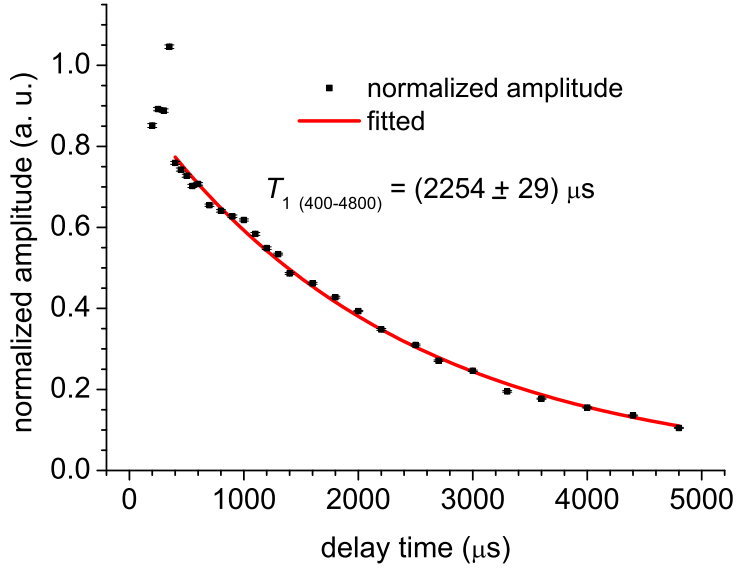


**Figure 4.** The square of the halfwidth of the absorption line profile as a function of the distance from the focal point of the focusing lens.

state for slowly varying probe field detuning  $\Delta$ . We have used the measured  $T_1$  and  $T_2$  times in the simulations. Details of the  $T_1$  measurement are given later in this section. In our experiment the scans have been repeated several times to reduce the measurement errors. The average of several scans is shown in figure 3 for  $t_d = 250\mu\text{s}$  and  $T = 9\text{K}$ .

In order to obtain a weak field limit for the pump (saturating) pulse we have done a Z-scan-type measurement. In figure 2 the lens focuses the modulated light beam into the sample. If the distance  $z$  between the sample and the focal point of the lens is larger than the Rayleigh-length, the atoms in the sample experience a field strength proportional to  $z^{-1}$ . Hence in Eq. (8) the peak depth of the spectral hole is proportional to  $z^{-2}$ : if we move the sample farther and farther from the focal point we obtain smaller and smaller saturation. On the other hand, the cross section of the focused beam is proportional to  $z^2$ . Therefore, by moving the sample along the optical axis the number of atoms involved in the process increases quadratically, while the signal coming from the atoms per unit area decreases quadratically, i.e. the intensity of the detected signal does not change. For our relatively low sensitivity detectors this is a great advantage. The result of the Z-scan measurement is shown in figure 4. By evaluating the measured half-widths as a function of  $z$  we obtained that in the limit of  $z \rightarrow \infty$  the half-width of the Lorentzian-distribution in Eq. (8) is  $\sigma = (42.7 \pm 0.7)\text{MHz}$  which yields a dipole relaxation time of  $T_2 \approx 90\text{ns}$ .

In Eq. (8) we can see that the peak depth of the spectral hole decays exponentially with a rate of  $T_1^{-1}$ . In a true two-level system this relation can be used to obtain the  $T_1$  population relaxation time. However, in case of  $\text{Er}^{3+}$ , there is the level  $^4\text{I}_{13/2}$  between the levels  $^4\text{I}_{11/2}$  and  $^4\text{I}_{15/2}$ . Hence there are two decay channels from the state  $^4\text{I}_{11/2}$  that we excite in our pump-probe experiment. Therefore, our model is a rough approximation of the real physical situation. Nevertheless, the line-shape function of Eq. (8) describes correctly the frequency dependence of the spectral hole, because the coherent pumping and probing do not create any coherence between the states  $^4\text{I}_{11/2} - ^4\text{I}_{13/2}$  or  $^4\text{I}_{15/2} - ^4\text{I}_{13/2}$ . However, the exponential dependence on the delay time  $t_d$  changes in



**Figure 5.** The decay of the peak depth of the spectral hole as a function of delay time between the pump and probe pulses.

a three-level model. In order to estimate the validity of our approximations in obtaining Eq. (7), we performed a series of measurements in which we measured the peak depth of the spectral hole as a function of delay time. The obtained curve is shown in figure 5. An exponential function fits very well the measured points, we obtained for the decay time constant  $T_1 = (2254 \pm 29)\mu\text{s}$ . This justifies our initial assumptions that  $T_2 \ll T_1$ .

#### 4. Summary

In this paper we have proposed an alternative technique to measure the  $T_2$  dipole relaxation time based on spectral hole burning spectroscopy. Our method is applicable if the atom can be treated as an effective two-level system interacting with a narrow linewidth coherent laser field. It is shown that for a sufficiently low intensity pump and probe fields, the half-width  $\sigma$  of the recorded absorption line-profile is related to the  $T_2$  time as  $T_2 = 4/\sigma$ . Based on the theoretical considerations, we have measured the dipole relaxation time associated with the transition  $^4I_{11/2} - ^4I_{15/2}$  of erbium in  $\text{LiNbO}_3:\text{Er}^{3+}$ . We plan to compare the measured  $T_2 \approx 90\text{ns}$  value with the result of a photon-echo measurement.

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